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PATENTS ACT, 1949
AMENDED SPECIFICATION

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PATENT SPECIFICATION

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DRAWINGS ATTACHED

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COMPLETE SPECIFICATION

A process for producing Crystallized Granules

We, NIPPON SHIRYO KOGYO CO., LTD., a body corporate organized under the laws of Japan, of 73, Shimonose, Tamiya-cho, Tokushima City, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following Statement:—

This invention relates to a process for producing crystalline granules of a crystalloid in a short cycle of operations and in high yield.

In accordance with the present invention there is provided a process for producing crystalline granules of a crystalloid, in which dextrose monohydrate is the main component but which may optionally contain impurities or added ingredients which give an improved product, which comprises producing an aqueous slurry consisting essentially of crystals of said crystalloid and saturated aqueous solution of said crystalloid, subjecting the aqueous slurry to a spray drying operation, the conditions being such that crystalline granules are formed containing about 13% by weight of moisture on a dry basis, which granules are composed of agglomerated crystals, the interstices of which granules contain supersaturated aqueous solution of said crystalloid, and subsequently gradually evaporating the free water content of each graule by ageing the granules in a stream of air from 3 to 5 hours.

It is necessary in the process of the invention to use a slurry which, when spray-dried forms granules which do not adhere one to another. The proportions of the two phases in the slurry are thus important. These proportions are such that in the granules immediately after the spray-drying described above, the crystal faces are in contact with each other through the supersaturated liquid phase. That is to say, the crystals come into contact with each other as the slurry is spray-dried to form granules composed of agglomerated crystals, the interstices of which granules contain supersaturated solution. The proportions of crystal phase and liquid phase in the slurry can be selected to ensure easy spray-drying.

An important feature of the spray-drying process of the present invention is that solid solutions should be avoided. In solid solution, the molecules exist in a random arrangement or orientation as in solution, but are closely packed so that they are prevented from being rearranged and from migrating to the points of crystal lattice.

In order to crystallise the liquid phase in the granules, more water than the minimum amount required for crystallisation must remain after spray-drying so that the liquid phase may enlarge the interstices among the granules in order to allow free movement of the molecules. This movement is accelerated as the temperature of the solution is increased. The temperature increase, however, could also re-dissolve or fuse the crystals already formed, and to avoid this effect, the temperature must be maintained within the limits in which the liquid phase remains supersaturated.

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The present invention thus consists of the following steps.

First, there is produced an aqueous slurry of a crystalloid consisting essentially of crystals of the crystalloid and saturated aqueous solution of the crystalloid. This slurry should be sufficiently free-flowing to be pumpable and, therefore, to be spray-dried efficiently. The slurry is then spray-dried so that the liquid phase is immediately concentrated. The precipitated granules are composed of agglomerated crystals, with the interstices containing supersaturated liquid. The extent of removal of water is restricted so that supersaturated liquid remains in the interstices of each granule. During and after the spray-drying, crystals grow in the supersaturated solution in the interstices of each granule. Finally the free water content of each granule is evaporated to form a dried product, which is ready for transportation and use. The above sequence of operations can be modified according to the impurities and additives present in the slurry. Since the supersaturated state is attainable by cooling as well as by concentration, it is possible to cool the slurry during spray-drying, in order to form granules in which the interstices contained supersaturated solution. However, since commercially, it is necessary to carry out the entire operation within a system held at a constant temperature, it is necessary to provide for the possibility of accidents and sudden temperature drops which might conglomerate the crystals. In the spray-drying of a highly concentrated solution of a crystalloid by a stream of cold air, there is always the possibility of accidental agglomeration.

It will be apparent from the above description that each of the granules obtainable by spray-drying consists of agglomerated crystals with supersaturated solution contained in the interstices between the crystals. That is to say, the crystals are glued together by the solution. On the other hand, the granules themselves are independent and free from the tendency to conglomerate or cake. Thus, the present invention is adaptable to the spraying of a crystalloid containing impurities, or of a crystalloid containing added ingredieents which give an improved product. The conception might appear contradictory when one considers the recrystallisation process by which products are purified. However, the actual value of any food or material is not always dependent on its purity, for there are many cases in which it may be more desirable to obtain less purified or mixed products. There are also many products, the actual value of which is not dependent upon their purity.

As a result of improvements in the hydrolysis of starch, it is now common to manufacture a solution in which more than 90% of the hydrolysate is pure dextrose. Substantially 100% hydrolysis of starch is now possible, and purification by crytallization is unnecessary.

For the most efficient utilisation of materials containing sugars, the total amount of sugar which can be obtained in a useful form is significant. The mother liquor remaining after recrystallisation is not a harmful producr, and can be sufficiently nutritive unless it is degraded by repeated operation. Extracts from plant juices have higher contents of sucrose than extracts from other sources, and it is doubtful therefore if there is any advantage to be gained by repeated recrystallisation of these extracts. There are some cases in which a product containing 3 to 5 percent of raffinose and other sugars may be satisfactorily utilised.

When a sugar which has been purified by crystallisation, is treated by the process of the present invention, it is converted into free-flowing granules which may be transported pneumatically, for example, the refined liquor from a sugar manufacturing plant, or the mother liquor after several cycles of sucrose operation, which contains 90% of sugar, may be made into a slurry, which may then be treated according to the process of the present invention to produce free-flowing granules without the formation of molasses. Furthermore, even the so-called soft sugar which is prepared by adding a small percentage of invert sugar to increase the sweetness, may also be treated according to the present invention to form free-flowing granules, which are readily soluble.

There has recently been a tendency to refrain from consuming too much sugar for health reasons and to use more synthetic sweeteners. However the latter leave some after-taste which is different from that of sugar. However, if sugar is mix-crystallised with a synthetic sweetener in accordance with the process of the present incention, a good tasting and sufficiently sweet product is obtained. Unlike a mixture of different types of powder, the granular product obtainable as described above is homogeneous. That is to say, the granules have a uniform composition and the distribution of the ingredients is not disturbed by screening or vibration.

The process of the present invention will be further described with reference to the accompanying drawings, in which:

Figure 1 is a diagram illustrating the changes in phase distribution in accordance with the process of the present invention; and

Figure 2 is a sketch ilustrating the spray-drying of a crystalloid in accordance with the process of the present invention; and

Figure 3 is a diagram illustrating the process described in Example 1 hereinafter.

Referring to the accompanying drawings, when a slurry which consists of saturated aqueous solution and crystals and which is

sufficiently fluid to be pumped, is spray-dried, spherical granules are formed as shown in (I) of Figure 2. This is because the liquid phase in which the crystals are suspended has a surface tension, which acts upon the crystals as the slurry is being spray-dried. Even a highly viscous concentrated solution, when in such a finely divided state, has an extremely large surface area per unit weight, and the evaporation of water from the liquid phase is rapid, so that the liquid phase shrinks in volume as it is concentrated.

In the process, the surface tension of the liquid phase causes the crystals to move inwardly towards the centre of each of the granules. When the sides or edges or the crystals come into contact with each other the outwardly exposed surfaces of each of the crystals grows, and these surfaces become dry. Thus the outer surface of the granules are prevented from adhering one to another. Thereafter, the liquid phase alone shrinks inwardly towards the centre of each granule. Due to surface tension and wetting phenomena, some solution remains surrounding the lines of contact between any two adjacent crystals, thus cementing the crystals together.

The granules formed by spray-drying fall in the state (II) of Figure 2 and as the granules fall, larger amounts of crystal surface are exposed. Centripetal movement of the liquid phase due to the surface tension results in granules which are prevented from adhering one to another due to the outer surfaces thereof being dry.

The degree of crystal growth may be ascertained by measuring the heat of crystallisation or the heat of crystal dissolution, and by quantitative partition analysis of water of crystallisation and free water, or by other suitable methods. It is quite easy to evaporate the negligible amounts of excess solvent after crystallisation, and this may be performed in any conventional manner.

Figure 1 shows, in sequence, the changes described above in respect of ingredients and phase distributions:

IVa, shows the pure product, and

IVb, shows the general composition of a product which contains some impurities or additives.

The present invention will be further described by means of the following Examples.

EXAMPLE 1

A starch hydrolysate liquid having a dextrose equivalent of 97 is prepared, and is then refined and evaporated to a density of Brix 67. The resulting concentrate is allowed to crystallise by slow agitation in a crystalliser at 20°C., into which it is preferable to add 0.5% by weight of dextrose monohydrate crystals or a material containing dextrose monohydrate as seeds of crystallisation. The time which is required for completion of the crystallisation, or which elapses before equilibrium is reached between the isolated crystals and the saturated solution at this temperature, is not constant but varies depending upon the degree of hydrolysis and purity of the liquor, although such an equilibrium is usually attained in about 8 hours. The process may be operated continuously by employing a suitable type of crystalliser, through which the concentrated liquor is allowed to crystallise. The material is charged into one side of the crystalliser and discharged from the other side through a narrow exit without the necessity of adding seeds of crystallisation.

The slurry thus obtained is composed of relatively small sized crystals and saturated solution, and is sufficiently fluid and stable to be pumped to a spray-drying chamber through a conduit. Therefore, the slurry can be fed continuously to the spraying apparatus. The slurry is spray-dried into small droplets or particles which are allowed to drop through a current of air in the chamber, the temperature of which is kept below 50°C., to obtain a product containing large amounts of dextrose monohydrate.

An example of the actual operation suitable for practical application on an economical scale is given below:

A drying chamber, consisting of a conical part having a length of 7.7 metres mounted on top of a cylindrical part having a length of 7.7 metres and a diameter of 7.7 metres, is equipped at the top with an atomising device 350 mm. in diameter, capable of rotating at 6500 r.p.m. to spray the slurry at a rate of 1000 kg/hr. A stream of air blown into the chamber is heated to 55°C., but the temperature drops to about 45°C., due to the heat of evaporation and the leakage of cold air. The granules deposited on the bottom of the chamber of the spray-dryer contain about 13% by weight of moisture on a dry basis, as the result of the evaporation of 230 kg. of free water from 1000 kg. of slurry. The granules have an appearance resembling melting snow flakes and are found to contain 65% of dextrose monohydrate crystals and 35% of solution when analysed. When the product is aged in a stream of air for 3—5 hours, it is completely crystallised and is sufficiently free-flowing to be transported by pneumatic means.

This process is illustrated in Figure 3 of the accompanying drawings.

It seems that greater efficiency may be obtained by more rapid drying, instead of drying in two steps. However, when dried too rapidly, the liquid phase in each granule does not crystallise into crystals but into a glass-like body which is hygroscopic and, hence, sticky. Thus, in order to obtain a desirable product, it is necessary that the moisture content of the granules be controlled during the growth of the crystals.

The ageing and simultaneous drying pro-

cess described above may be operated continuously by means of a suitable apparatus, for example, a rotary or belt conveyor, or by pneumatic equipment in which the temperature, the relative humidity, and the velocity of the air stream are controlled.

Example 2

Dextrose monhydrate crystals were obtained by centrifuging a slurry of glucose which has a purity greater than 99%, and which contains a small percentage of oligosaccharides. As a solution of these wet crystals has a high crystallinity, it is suitable for use in the process of this invention. The crystals are partially dissolved in a requisite amount of water, or are admixed with a saturated solution of dextrose monohydrate, so as to prepare a slurry which contains approximately equal amounts of dextrose monhydrate crystals, dextrose in solution, and water. The slurry thus prepared is not economical for use in the production of dextrose by the process of the present invention, but is suitable for use in the production of homogeneous products which contain additives and in which dextrose monohydrate is the main component.

Example 3

A starch hydrolysate liquor having a dextrose equivalent of 97 is refined and concentrated to a density of Brix 67, and is then allowed to crystallise by the method described in Example 1. During the preparation of the slurry a synthetic sweetener agent, for example, 1—1.5% of sodium cyclohexyl sulphamic acid as a 10% solution is added. The sweetening agent does not interfere with the crystallisation of the dextrose and dissolves homogeneously in the liquid phase of the slurry. The slurry is pumped to the spray-drier, sprayed, aged and dried in the manner described in Example 1, giving a non-caking product having the same appearance and as much calorific value as the product of Example 1, but being sweeter because of the sweetening agent homogeneously distributed therethrough.

Example 4

The slurry prepared as described in Example 2 is suitable for use in the process described in Example 3. Thus, when the slurry is admixed with a synthetic sweetening agent, and the slurry spray-dried, there are obtained homogeneous, sweetened granules similar to those described in Example 3.

Example 5

A slurry prepared as described in Example 3 or Example 4, is admixed with an organic acid instead of a synthetic sweetening agent. The slurry is spray-dried in the manner described in Example 1 to obtain a granular product which contains the organic acid uniformly distributed between or on the dextrose monohydrate crystals of each granule. Solid organic acids, for example, citric acid, succinic acid or tartaric acid can be used in this process.

Example 6

A starch hydrolysate liquor having a dextrose equivalent of 96 is refined and concentrated while regulating its density, and is then crystallised into a slurry similar to that described in Example 1. Components of various drinks or foods, for example, concentrated fruit juices, vegetable juices, coffee extracts, or cacao powder, can be successively admixed homogeneously with the slurry, prepared according to Example 2. Spray-drying and subsequent elimination of residual moisture in the manner described in Example 1 results in a product which is readily soluble and easily dispersible because of the homogeneous distribution of the components in the individual granules.

It is preferable when the slurry contains such addition ingredients as described above, to control the fluidity of the slurry by regulating its water content, since the presence of high molecular weight materials increases the viscosity of the slurry.

Instant orange juice is prepared by means of the process described in Example 1, by admixing the following components in the proportions indicated.

Glucose slurry	100 kg. (as solid)
Concentrated orange juice	80kg.
Sodium cyclohexylsulphamate	10kg.
Soluble saccharine	3kg.
Orange oil	15kg.
Citric acid	35kg.

WHAT WE CLAIM IS:—

1. A process for producing crystalline granules of a crystalloid, in which dextrose monohydrate is the main component but which may optionally contain impurities or added ingredients which give an improved product, which comprises producing an aqueous slurry consisting essentially of crystals of said crystalloid and saturated aqueous solution of said crystalloid, subjecting the aqueous slurry to a spray drying operation, the conditions being such that crystalline granules are formed containing about 13% by weight of moisture on a dry basis, which granules are composed of agglomerated crystals, the interstices of which granules contain supersaturated aqueous solution of said crystalloid, and subsequently gradually evaporating the free water content of each granule by ageing the granules in a stream of air from 3 to 5 hours.

2. A process as claimed in claim 1, in which the slurry contains a synthetic sweetening

agent, an organic acid and/or a colouring agent.

3. A process as claimed in claim 1 or claim 2, in which the slurry contains a fruit juice, a vegetable juice, a coffe extract and/or cacao powder.

4. A process for producing crystalline granules of a crystalloid as claimed in claim 1 and substantially as hereinbefore described with reference to any one of Examples 1 and 3 to 6.

5. Crystalline granules of a crystalloid whenever prepared by the process claimed in any one of the preceding claims.

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FIG.1

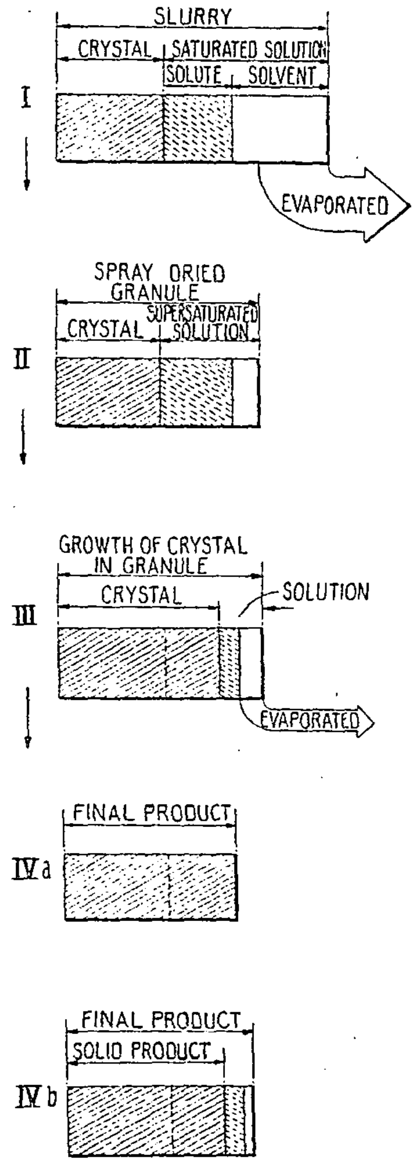
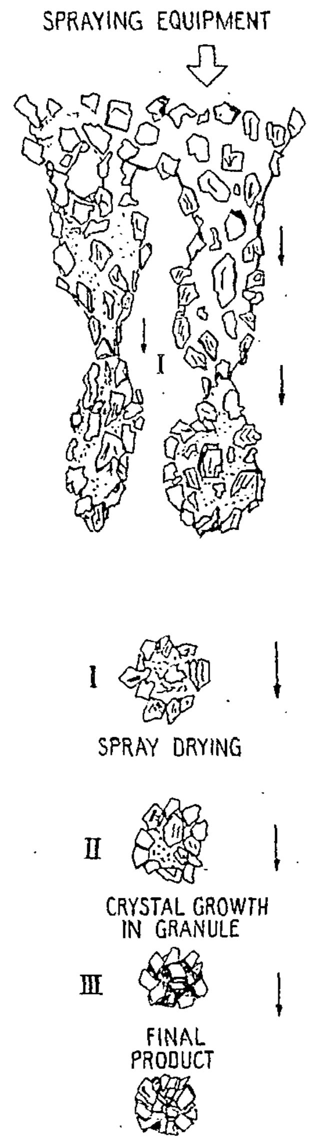

SLURRY
CRYSTAL
SATURATED SOLUTION
SOLUTE
SOLVENT
I
EVAPORATED
SPRAY DRIED GRANULE
CRYSTAL
SUPERSATURATED SOLUTION
II
GROWTH OF CRYSTAL IN GRANULE
CRYSTAL
SOLUTION
III
EVAPORATED
FINAL PRODUCT
IVa
FINAL PRODUCT
SOLID PRODUCT
IVb

FIG.2


SPRAYING EQUIPMENT
I
I
SPRAY DRYING
II
CRYSTAL GROWTH IN GRANULE
III
FINAL PRODUCT

1112553 AMENDED SPECIFICATION
2 SHEETS *This drawing is a reproduction of the Original on a reduced scale*
Sheet 2

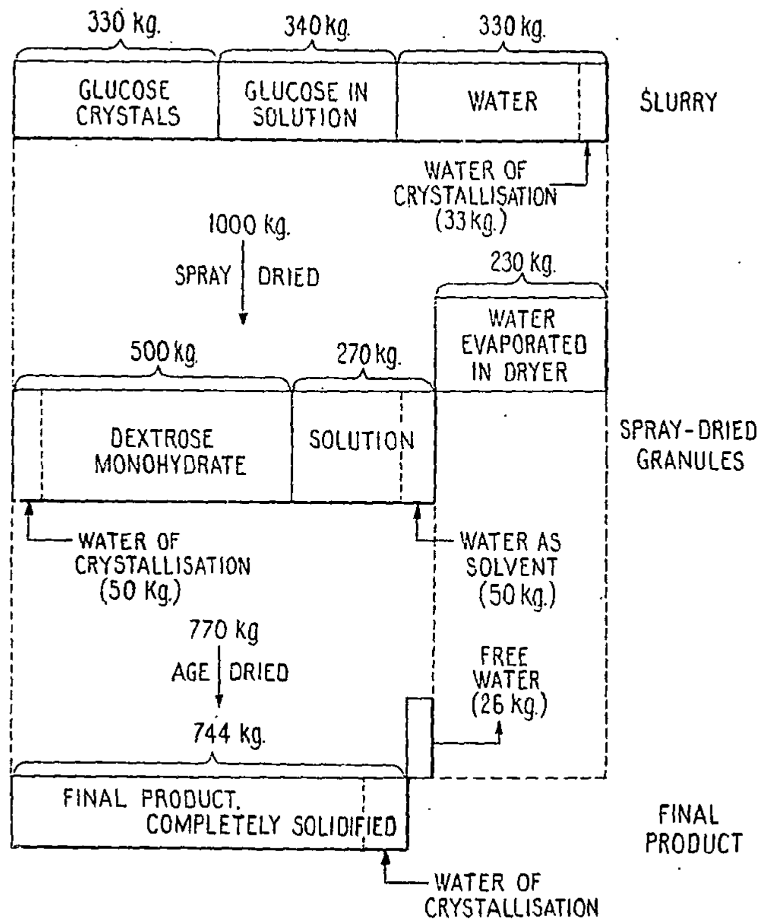


FIG. 3